Studies of Tritium Characterization in Concrete at the Savannah River Site

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Cementitious Materials for Waste Treatment, Disposal, Remediation and Decommissioning Conference

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Outline

- Scope of SRS Tritium in Concrete Characterization
- Challenge of Tritium Detection
- Literature- Exists!
- Current Study
  - Tritium Characterization with Depth at Several SRS Locations
  - New Drill/Leaching Techniques Developed
  - Surface Characterization Methods- Comparison with Bulk

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SRS Needs for Tritium Characterization in Concrete

- Nuclear Materials Production (^{239}Pu, ^{238}Pu, ^{3}H, others)
  - 5 Heavy Water Moderated Production Reactors- Deactivated
    - Tritium Produced as By-Product in Moderator Water from Neutron Irradiation
  - Heavy Water Processing (Purification) Facilities
  - Legacy Heavy Water Storage Ongoing
- Tritium Facilities
  - Original Tritium Facility: 232-F
    - Operated 1955-1958
    - Dismantled Mid 1990's- "Green Fielded"
    - Required Extensive Core Drilling for Tritium Characterization- Not Part of Original Project Scope
  - Building 232-H Deactivation Complete 2005- "Cold, Dark, Dry", No Concrete Characterization
  - H Area Old Manufacturing Building (HAOM)- Deactivation and Renovation/Reuse

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Tritium Detection Challenging

- ^{3}H Weak Beta Emitter: 5.7 keV Average, 18.5 keV Maximum
  - Penetration ~ 6mm Air, ~6 micron in Material with Unit Density
  - Almost Any Penetration in Material- Tritium Becomes Undetectable
- Readily Exchanges With ^{1}H in All Substances- Water is Ubiquitous in Concrete: $^{1}H^{3}HO$ a.k.a. HTO
- Small Beta Penetration Is the Challenge- Compare with Detection Gamma Emission

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Tritium In Concrete Literature

- Numata et.al- Thermogravimetric Study (S. Numata et al., "Tritium Inventory in Portland Cement Exposed to Tritiated Water Vapor," Journal of Nuclear Materials 171, pp. 350-359, 1990)
  - Pore & Capillary Water 67%
  - Water of Crystallization 20%
  - Constituent Water in Calcium Hyroxide + Calcium Silicate Hydrate 13%
- Kraznai- CANDU Reactor Decommissioning- Tritium Resides Throughout Concrete (J. P. Krasznai, "The Radiochemical Characterization of Regular- and High-Density Concrete from a Decommissioned Reactor," Waste Management, Vol. 13, pp. 131-140, (1993)
- Review (Canada): R. S. Dickson, "Tritium Interactions with Steel and Construction Materials in Fusion Devices – A Literature Review," Report AECL-10208, CFFTP-G-9039, Chalk River Laboratories, November, 1990.
- Others…

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- Simplified Hammer Drill Method to Extract Concrete Samples with Depth
- Nitric Acid Dissolution Technique- Enables LSC of Samples to Characterize Tritium
- Choose Locations in Facilities Likely to Have Tritium in Concrete for Study
- Relate Bulk to Surface Activity as Measured by Two New Detectors

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Hammer Drill Method for Sample Collection at Depth

- Hand-Held Hammer Drill- 16mm or 19mm Bits
- 1” Depth Increments
- Collect Powder in Vial, Vacuum Remaining Dust
- Plenty of Material Extracted- 20 g per Increment
- Worked Well in All Cases

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New Leaching Method

- Leach 1 g of Depth Sample in 10 mL of 1N HNO_3 Overnight
- Distill ~2 ml of Leachate
- Analyze 2 ml aliquot of Distillate in 18 ml LSC Cocktail for Tritium
- Compares Well with Flowing Air Extraction at 550° C./Water Collect/LSC

Calcium Analysis Also Performed on All Samples- Correlation of Tritium With Calcium- Sodium Peroxide Fusion, Ca, Si Analysis by ICP-MS

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Heavy Water Processing Area 420-D

Same Data- Normalized to Ca Concentration

Figure 1. H-3 Concentration Per Gram of Sample at 420-D

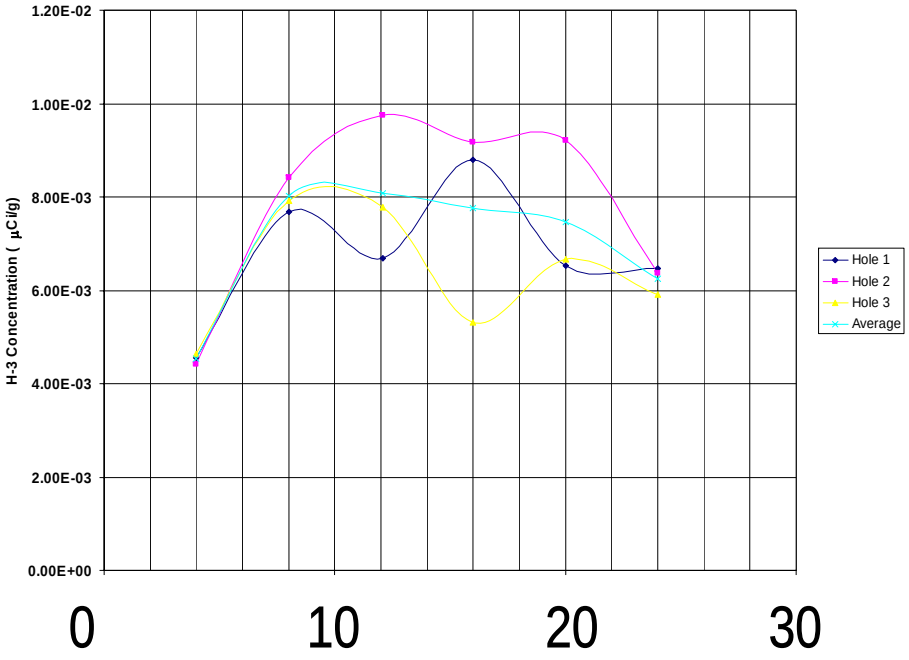
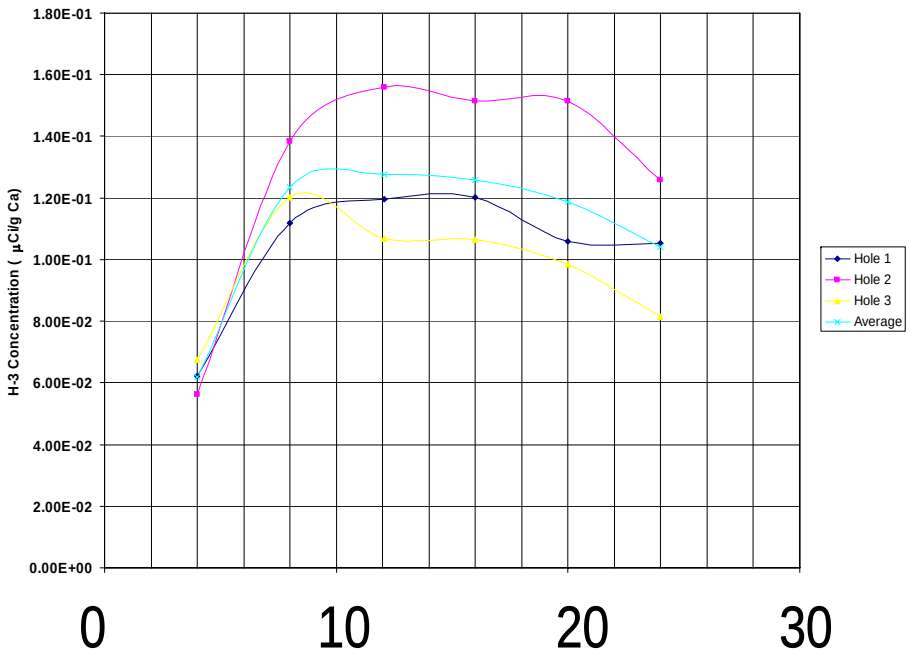


Figure 2. H-3 Concentration Per Gram of Calcium at 420-D



Sporadic Exposure to Tritiated Heavy Water- eg. Spills

Flat Concentration Profile in Bulk, Weathering Reduces Surface

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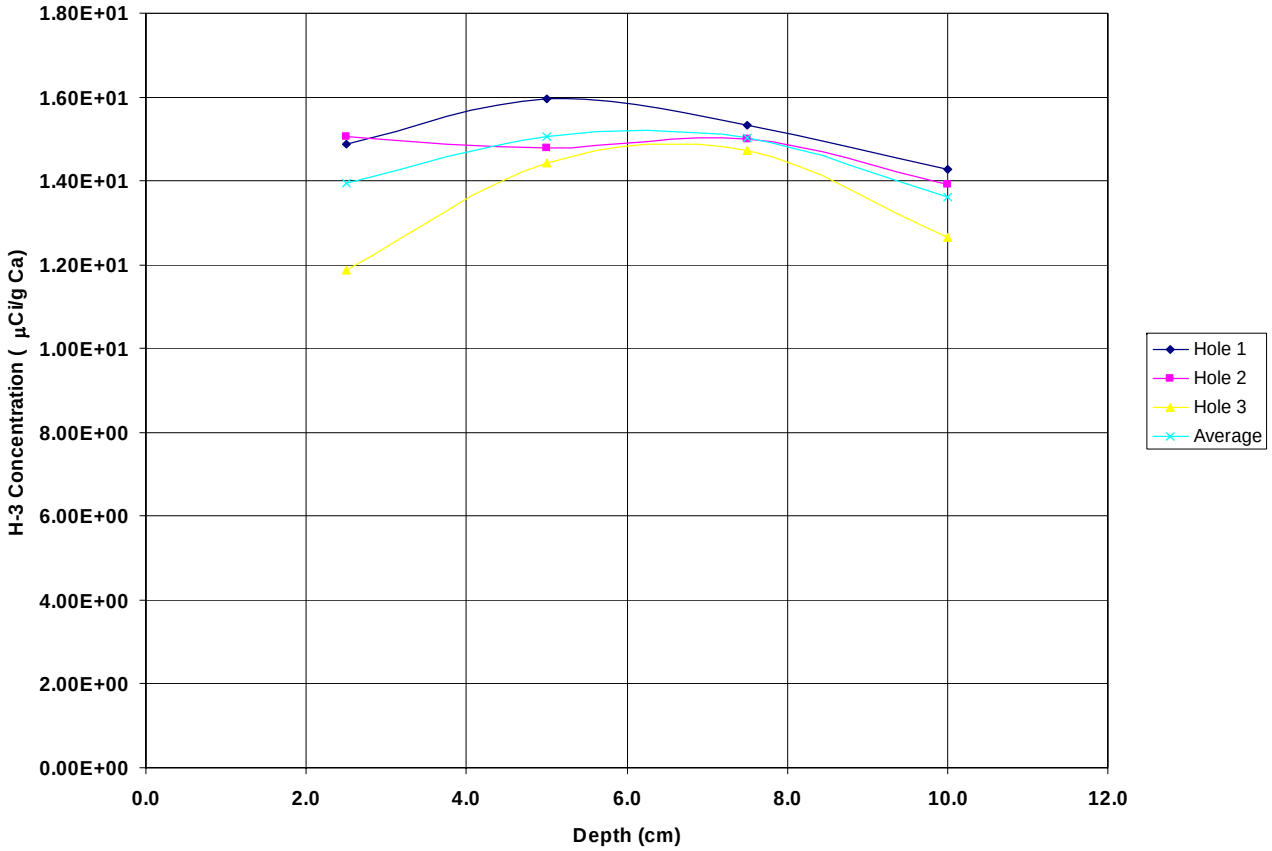
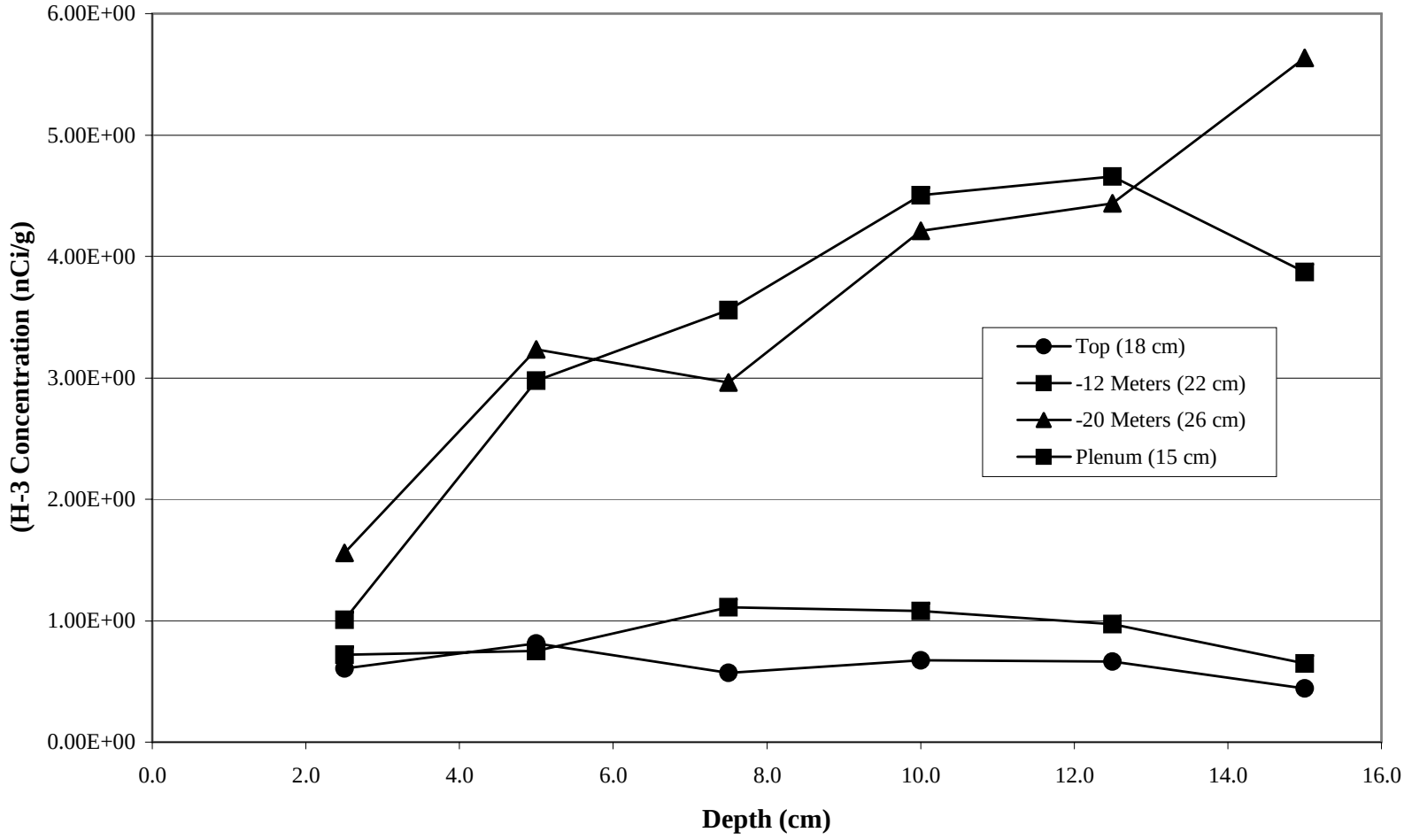
H Area Old Manufacturing



Location Highly Surface Contaminated- Near Machining Equipment- Oil?

K Reactor Ventilation Stack

Variation of Tritium with Depth- Vapor Exposure!



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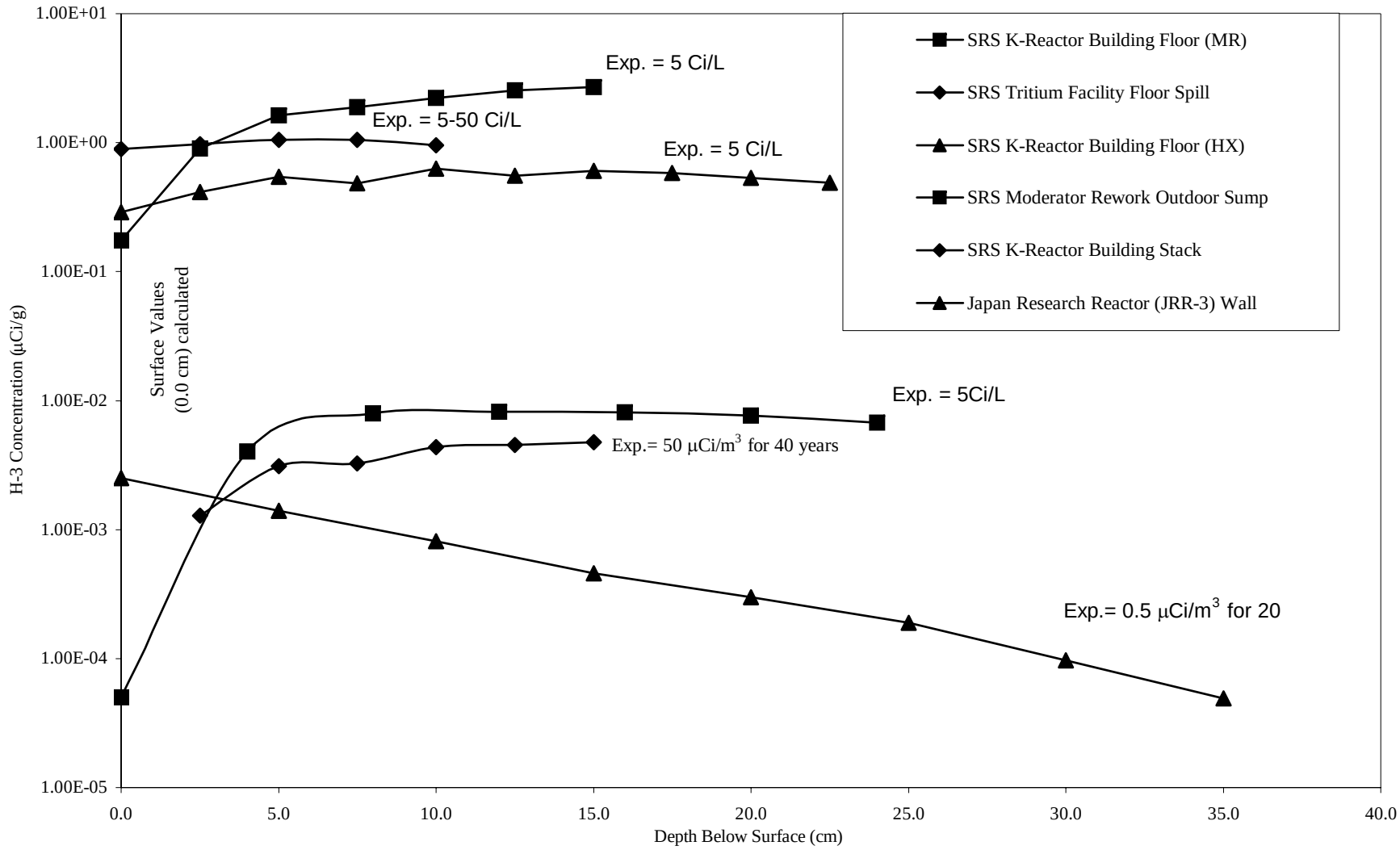
Comparing Tritium Concentration Profiles



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Surface Detection Methods Studied

- SAM- Surface Area Monitor (Ontario Hydro Labs/Qualprotech)- Electrometric Tritium Detection at a Surface
- E-Perm System (Rad Elec)- Discharge of a Pre-charged PTFE (Teflon®) Disk (“Electret”), Voltage Reduction of Electret Proportional to Radiation at Disk

Electret

Reader

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Surface Activity Measurements

Study Area	Bulk Mass Activity (µCi/g)	Correlation Factor (g/cm^2)	Surface Activity Calculated (nCi/cm^2)	Surface Activity Measured (nCi/cm^2)
420-D	7.0 E-03	1.2E-02	0.8E-01	0.2 - 4.1 E-01
HAOM	1.0 E+00	1.4E-02	1.4E+01	1.4 E+01

SAM and E-Perm Resulted in Similar Values of Surface Activity!

Constant Correlation Factor- Attributed to

- Liquid Exposure in Both Cases
- Porosity of Concrete- Allows Collection of More Decays than Flat Surface

Surface Smears- Varying Collection Efficiency (5% HAOM, 0.1% 420-D)

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Summary

- Exposure of Concrete to Tritiated Liquid- Tritium Uptake Through Bulk, Relatively Constant Concentration with Depth, Weathering at Surface
- Exposure of Concrete to Vapor HTO ($^3H_2$ as well?)- Diffusive Type Variation from Exposed Surface Inward
- Simple, Cheap Hammer Drill & Nitric Acid Leaching Methods Demonstrated for Characterization
- Surface Activity Measured with SAM, Electret Correlates with Bulk- Surface Smears Cannot Be Used to Estimate Bulk Concentration
- ASSUME TRITIUM PENETRATES THROUGHOUT CONCRETE!

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Publications

- R.C. Hochel, E.A. Clark. “Tritium Characterization in Cement and Concrete”, Radioactive Waste Management and Environmental Restoration, Vol. 22 (3) pp. 175-204 (2000).
- R.C. Hochel, E.A. Clark. “Corroborative Studies of Tritium Characterization and Depth Profiles in Concrete”, Radioactive Waste Management and Environmental Restoration, Vol. 22 (4) pp. 394-419 (2002).

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